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Experimental analysis of the retention behavior of shales

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ABSTRACT: Water retention mechanism has a primary importance in either fluid trapping due to the capillary forces or in the resaturation of shale formations after desaturation. This paper illustrates the advances in the experimental analysis of the water retention behaviour of shales under non-isochoric conditions which have recently been achieved by the authors. The testing methodologies, which allow for the determination of the main drying and wetting paths, involve the direct control of the shale water content and the subsequent measurement of the total suction at equilibrium by a psychrometer. Furthermore, the volume change response of the material upon total suction variations is investigated: a fluid displacement technique with a non-polar liquid is used in order to assess the volume changes of the shale samples and to compute the degree of saturation. Selected results are presented for two Mesozoic shales from the northern region of Switzerland: the Opalinus Clay from Mont Terri and the so called “Brown Dogger” from the Schlattingen site in the Molasse Basin.

1. INTRODUCTION

Growing interest toward the geomechanical behaviour of shales is nowadays observed mainly due to the fields of application in which this material has been involved in the recent years such as petroleum industries, nuclear waste storage and sequestration of CO₂. In any such application, a deep understanding of the hydro-mechanical behaviour of shales is of primary significance. The water retention mechanisms play a major role in either fluid trapping due to the capillary forces present in low permeability formations or in the resaturation of shale formations after desiccation [1]. This paper presents the advanced experimental techniques that have been recently developed by the authors for testing the water retention behaviour of shales. Testing procedures are conceived with the aim to create a standard protocol to tackle the determination of the wetting and of the drying paths of the shale water retention behaviour. The methodologies are based on the direct control of the shale water content and on the subsequent measurement of the total suction at equilibrium by a psychrometer. The swelling/shrinkage of shale is related to suction, or the degree of saturation variations; in order to investigate the volume change response of the material upon total suction variations a fluid displacement technique with a non-polar liquid is

used. The technique allows to assess the changes in void ratio of the shale specimens and to compute the degree of saturation.

2. TESTED SHALES

The two Mesozoic shales considered in the present study come from the northern region of Switzerland: the Opalinus Clay sample was obtained from the Mont Terri Underground Laboratory and the “Brown Dogger”, from a deep geothermal well (depth of 766.67 and 778.30 m) near the village of Schlattingen in the Molasse Basin. The tested Opalinus Clay (OPA) was recovered in the Gallery 98 of the Mont Terri URL from the shaly facies, consisting of dark grey silty, calcareous shales with a typical clay content of 55-60%, 25-30% carbonate, 5-10 % quartz and 10-15% biotritus. The core samples of “Brown Dogger” (BD) originated from the Variscan- and Parkinsoni-Würtembergica beds (Bathonian/Bajocian age), consisting of silty to clay-rich marls with a clay content of 25-45%, 30-50% carbonate and 20-25% quartz [1].

The geotechnical characterization consists in the determination of the particle density (ρ_s), the bulk density (ρ), the water content (w), the void ratio (e), the degree of saturation (S_r) and the Atterberg limits (the

liquid limit w_L and the plastic limit w_P). The results of the geotechnical identification of the cores are reported in Table 1:

Table 1: Geotechnical characterization of the tested shales

Shale	OPA	BD
ρ_s (Mg/m ³)	2.74	2.72
ρ (Mg/m ³)	2.46	2.55
w (%)	6.9	2.7 - 3.5
e (-)	0.21	0.09 - 0.11
S_r (%)	92	78 - 92
w_L (%)	38	25 - 29
w_P (%)	23	23

Figure 1 depicts the grain size distributions of the two considered shales. The grain size distributions are obtained by sedimentation analysis following the procedure described as follows: the material is first crushed using a grinder, and the fraction passing the ASTM No. 35 sieve (with an aperture diameter $d = 500 \mu\text{m}$) is selected. The crushed material is placed in distilled water along with a dispersing agent and is then shaken overnight. From the obtained curves in Figure 1 it is possible to observe that they are situated on the left side of the maximum dimension of the selected material, i.e. the material does not present particles with a diameter larger than the maximum dimension of the crushed blocks; the previous observation validates the selected procedure for preparation of the sedimentation analysis [1].

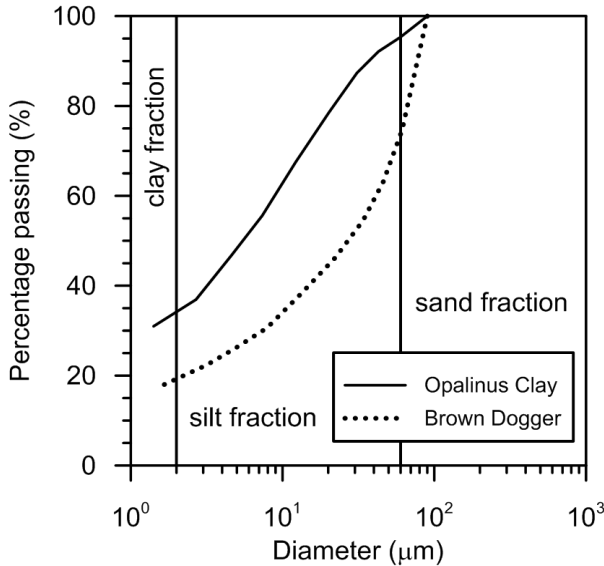


Fig. 1. Grain size distribution of the tested shales.

The pore size density (PSD) function of the tested shale cores is determined through the Mercury Intrusion Porosimetry (MIP); this technique consists in forcing the penetration of mercury into a sample and in measuring the intruded volume of mercury as a function of the applied pressure. The required pressure is inversely proportional to the size of the progressively filled pores.

The obtained results are presented in Figure 2. The PSD functions are found to be unimodal for both tested shales; they show pore modes at approximately 20 nm and 15 nm for the OPA and the BD samples, respectively.

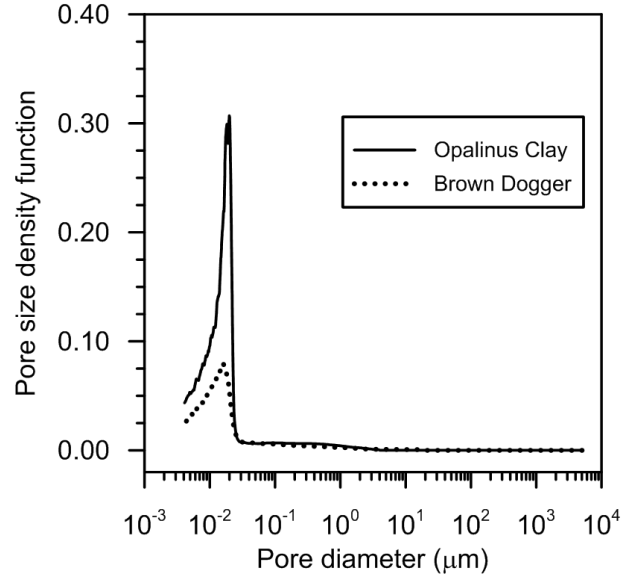


Fig. 2. Pore size density function of the tested shales.

The total cumulated void ratios obtained with the MIP tests are 0.13 for the OPA sample and 0.074 for the BD sample. Nevertheless the MIP technique does not allow the investigation of the overall porosity since no complete intrusion can be achieved; this limit can be related to pores on the surface of the samples that are filled with mercury before the pressure increments started and to the smallest pores that require higher pressures to be intruded (minimum pore diameter that can be intruded with the used apparatus was 3.8 nm). This observation can be confirmed by the comparison of the cumulated void ratios obtained with the MIP and the corresponding values reported in table 1.

3. TESTING METHODOLOGIES

This section describes the experimental methodologies established to analyse the water retention behaviour of shales under unstressed conditions. In all the depicted testing procedures synthetic waters are used with the aim to reproduce the in-situ pore water composition [2, 3]. The osmotic suctions of the synthetic waters used are measured by a dew-point psychrometer and result in values of 1.2 MPa for the OPA sample and 0.98 MPa for the BD sample.

As already introduced the presented testing techniques are based on the direct control of the water content and the subsequent measurement of the suction value. The imposed water content is applied differently for the wetting and drying paths.

The total suction readings are carried out by means of a dew-point psychrometer (Decagon, WP4c) [4, 5]. The apparatus gives indirect measures of the total suction of a sample by reading the relative humidity established by the sample in a closed environment. The chilled-mirror dew point technique is used to measure the relative humidity. The total suction is then obtained by the psychrometric law [6] which relates the relative humidity (RH) and the absolute temperature (T) to the total suction (ψ):

$$\psi = -\frac{\rho_w RT}{M_w} \ln(RH) \quad (1)$$

where R is the universal gas constant and ρ_w and M_w are the density and the molecular mass of water, respectively.

The first step of the testing procedure requires the cutting of slices measuring 7 – 8 mm in height from the shale core; one slice is divided into five parts and each part is then divided into three specimens of about 2x2x1 cm³. The three central specimens are immediately tested by means of the WP4c in order to obtain the initial conditions of the material. After the determination of the total suction, the volume of the specimens is measured. Indeed, the computation of the degree of saturation requires the measurement of the volume of the shale samples at each equilibrated state.

The method adopted for the volume measurement is based on a fluid displacement technique which uses pycnometers filled with kerdane, a paraffin oil used for its immiscibility with water and its ability to invade the air-filled pore spaces in the surface without affecting the soil structure [7]. After the determination of the initial conditions of the tested shales, the main wetting and drying paths can be determined.

3.1. Main drying path

The determination of the main drying path requires the initial saturation of the core slices; saturation is achieved through the use of a sealed-glass jar filled by synthetic water. The slices are placed on filter papers, which in turn are placed on a porous stone; the latter is immersed in the synthetic water thus the material is wetted by capillary action allowing for volume changes of the samples. In order to assess the equalization of internal humidity, the weight evolution over time is monitored. Nearly two weeks are required for the slices to equalize. The wetted slices are then divided in small parts measuring approximately 20 x 20 x 8 mm³. Each part is placed in a desiccator and dried for the amount of time necessary to achieve the target water content of that particular part. After the drying step, the samples are packed for at least three days to allow for the internal redistribution of water.

Once the internal equilibrium is reached, the total suction is measured using the dewpoint psychrometer. Subsequently, the volume measurement of the sample is performed through the technique previously introduced. In the end, the water content, void ratio and degree of saturation can be determined.

3.2. Main wetting path

In order to reproduce the main wetting path, the shale samples have to reach an initial dry state. This is obtained by placing the core slices in a desiccator containing silica gel, the material is left to dry for approximately three weeks (applied total suction of approximately 270 MPa) until no significant change in their weight is detected. The slices are then cut in pieces and a certain amount of synthetic water is added to each part to obtain the target water content. Each part is then placed in a hermetic container and let to reach internal equilibrium of relative humidity. The total suction is then measured through the dewpoint psychrometer.

The last step consists in the measurement of the sample volume thus the water content, the void ratio and the degree of saturation of the considered shale sample can be evaluated and related to the measured total suction.

4. RESULTS

The water retention behaviour of OPA is reported in figures 3, 4 and 5 in term of water content, void ratio and degree of saturation versus total suction, while the retention behaviour of the BD core samples are presented in figures 6, 7 and 8. The points reported in the graphs represent a measurement which corresponds to the average of three specimens, each of which followed an identical drying/wetting path. The initial state of each sample is also indicated.

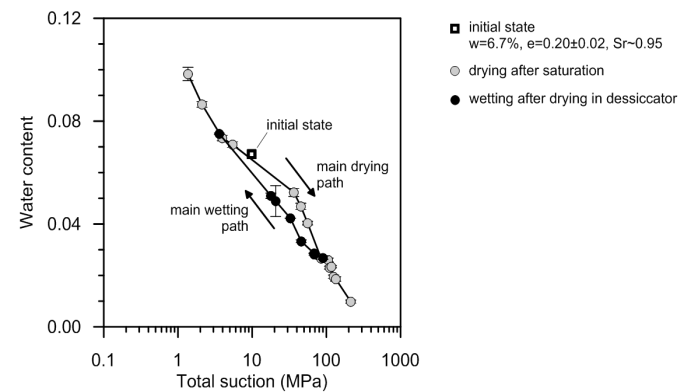


Fig. 3. Water retention behaviour of the OPA core sample: the water content as a function of the total suction.

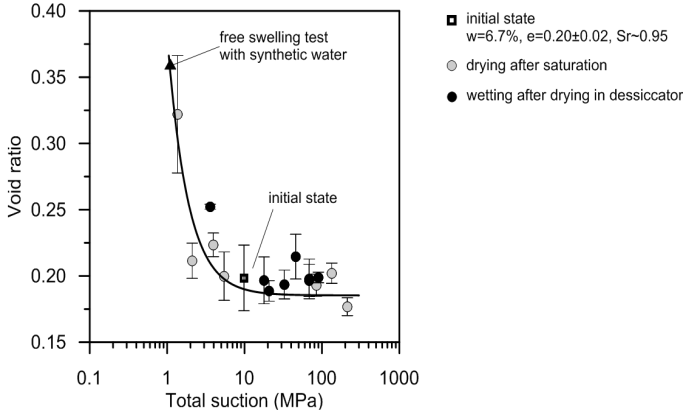


Fig. 4. Water retention behaviour of the OPA core sample: the void ratio as a function of the total suction.

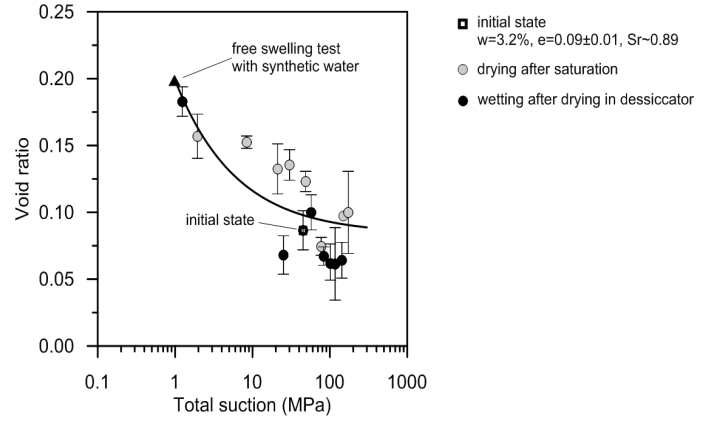


Fig. 7. Water retention behaviour of the BD core sample: the void ratio as a function of the total suction

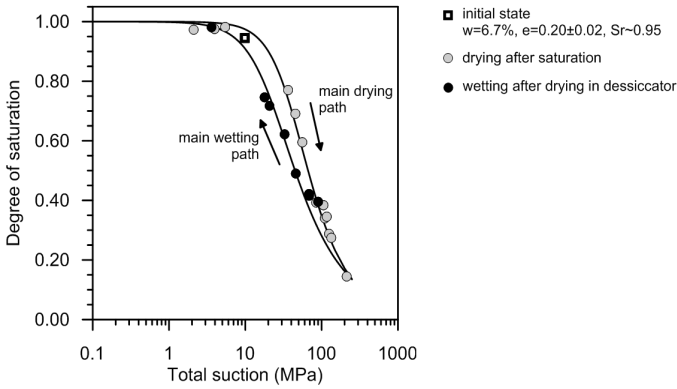


Fig. 5. Water retention behaviour of the OPA core sample: the degree of saturation as a function of the total suction.

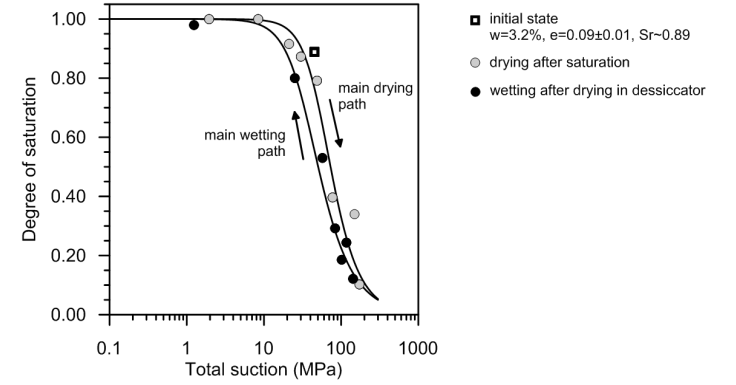


Fig. 8. Water retention behaviour of the BD core sample: the degree of saturation as a function of the total suction

Both the considered materials appear to have an initial degree of saturation close to one, this observation is in agreement with the fact that both materials presented saturated in-situ conditions. The initial state is found for both the materials along the main drying paths (figure 5 and 8); this observation is related to the effect of the desaturation which is a consequence of the coring process and of the exposure to atmosphere before preservation.

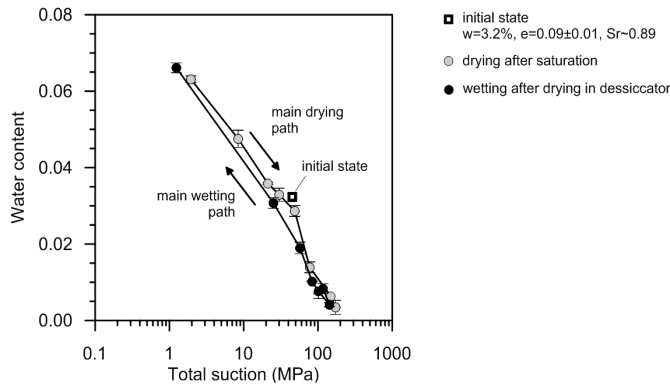


Fig.6. Water retention behaviour of the BD core sample: the water content as a function of the total suction

Figures 3 and 5 for the OPA sample and figures 6 and 8 for the BD sample show that the main drying and wetting paths are well distinguished and hysteresis zones can be observed. The evolution of the void ratios with the total suction (figures 4 for the OPA sample and 7 for the BD sample) shows that the porosity changes are more significant for the lowest range of suction variations. The void ratio corresponding to the lowest measured total suctions are in very good agreement with the results of the free swelling tests which have been carried out with the same synthetic waters used for determining the water retention curves (figures 4 and 7). The shrinkage limits (assumed to be defined by $\Delta e/\Delta \psi < 0.1\%$) are identified at suction values of approximately 10 MPa for the OPA and 15 MPa for the BD. The corresponding water content values read along the main drying paths are 6.4% and 4.0% for the OPA and the BD, respectively. The data of the void ratio distribution versus total suction are fitted with the following incremental expression:

$$\dot{e} = -\frac{\dot{\psi}}{K_{ref}} \left(\frac{\psi_{ref}}{\psi} \right)^m (1 + e_0) \quad (2)$$

in which K_{ref} , ψ_{ref} and m are the fitting parameters. The obtained values of the parameters are reported in table 2.

The retention curves in terms of degree of saturation versus total suction are depicted in figures 5 and 8. The experimental points are fitted using a van Genuchten's expression [8]:

$$S_r = \left(1 + \left(\frac{\psi}{P} \right)^n \right)^{\left(\frac{1}{n} - 1 \right)} \quad (3)$$

where P and n are the fitting parameters; the obtained values are reported in table 2. The curves show an air entry value (gas entry pressure) of approximately 13 MPa for the OPA core sample and 22 MPa for the BD core sample (the values computed for a degree of saturation of 95%). The higher air entry value of the BD core sample is in agreement with the lower porosity (Table 2).

Table 2: The fitting parameters for the water retention behaviour.

Shale	OPA	BD
K_{ref} (MPa)	14.6	22.0
ψ_{ref} (MPa)	1.32	1.10
m (-)	2.63	1.55
n wetting (-)	1.82	2.4
n drying (-)	2.1	2.8
P wetting (MPa)	22.0	35.9
P drying (MPa)	41.0	57.2

5. CONCLUSIONS

The paper presented the experimental techniques that have been developed by the authors for the analysis of the water retention behaviour of shales. Selected experimental results for two shales from the northern region of Switzerland have been presented and discussed.

The elaborated testing procedure for the determination of the water retention curves of shales consisted in controlling the water content by the addition of synthetic water or by the desiccation of the material in a desiccator. Total suction measurements are performed through the use of a dew-point psychrometer. After every suction measurement, the sample volume changes along the wetting and drying paths are systematically assessed; thus the computation of the degree of saturation and the evaluation of the shrinkage limit of each material can be achieved.

In order to observe significant variations in the degree of saturation a wide range of suction had to be considered due to the low porosity of the tested shales. The method is proved to be suitable for describing the water retention behaviour along the main drying and wetting paths and

for highlighting the hysteretic behaviours of the retention curves of shales.

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